A Microwave-Ultrasonic cell for Sound Speed Measurements in Liquids[1]

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ABSTRACT

In this work we describe the preliminary results obtained in developing a prototype cylindrical ultrasonic cell designed to apply a microwave resonance technique to the determination of its internal dimensions. Since the apparatus is intended for speed of sound measurements in pressurised liquid phase mediums, the cell design is such that a double reflector pulse-echo technique can be used for time of flight measurement. The main absorption and dispersion effects which affect the acoustic measurement in this interferometer-like configuration and the entity of the corresponding corrections are considered and discussed. The performance of the experimental apparatus and method in terms of achievable precision and accuracy was tested by measuring the speed of sound in water on a single isotherm at 325 K between 0.1 and 90 MPa.

KEY WORDS: speed of sound, microwave resonator, pulse-echo method.

1. INTRODUCTION

Any sound speed measurement apparatus needs an independent procedure to evaluate some geometric feature of the system, directly or indirectly related to the physical path length travelled by the sound wave: in steady-state techniques, these are the characteristic dimensions of a resonant cavity of simple geometry; in transient techniques, the distance between a pulse source and a reflector. While high accuracy and precision can easily be achieved in time or frequency measurement, the determination of the actual geometric path length appears to be the main limiting factor when attempting to reduce the uncertainty budget of a speed of sound measurement.

In this work, we describe a prototype ultrasonic cell consisting of a plane PZT transducer and stainless steel reflectors and spacers which form a double cylindrical conducting cavity. The transducer-reflector distances are calculated determining the length of the two cavities from measurements under vacuum of microwave resonances of a few TE and TM modes of different symmetry. Corrections are then applied on the basis of the coefficient of compressibility of stainless steel to keep into account the pressure dependence of the calculated path length difference.

In the acoustic configuration, the cell is designed to determine the difference in the propagation time of an acoustic pulse between the PZT transducer and the opposite reflectors, realizing a traditional double pulse-echo method [1, 2]. As the acoustic source and the propagating cavity have the same section, the acoustic propagation is very much like that taking place in an interferometer. The entity of the correction which accounts for the absorption effect from the walls of the two cavities results to be in the order of a few ppm, with a negligible additional contribution to the overall uncertainty.

Shape distortion of the acoustic pulses as a consequence of the guided-wave propagation modes was not modeled, however we did not have any significant experimental evidence of such an effect.

As a first test of the performance of the method, pulse-echo measurements were performed at 325 K between atmospheric pressure and 90 MPa on a sample of high purity water and the results combined with dimensional measurements obtained measuring at the same temperature the microwave resonances of the TE111 and TE112 modes of the two cylindrical cavities. The determined speed of sound data have been compared with the prediction of the IAPWS-95 formulation [3] with an observed deviations of 0.05%.

1. MICROWAVE MEASUREMENTS

1.1 Experimental apparatus

The prototype microwave-ultrasonic cell used in this work is shown in Fig. 1.

Fig. 1. Photograph of the double cylindrical cavity used in this work for microwave and pulse-echo measurements

It consists of two AISI 303 stainless steel cylindrical cavities having an outer diameter of 60 mm, an internal diameter of 42 mm and a length of 40 and 60 mm respectively. Two solid stainless steel reflectors (4 mm thick) are fixed to the bases of the cell as to form a couple of geometrically regular cylindrical conducting cavities when completed by a piezoelectric disc having a diameter of 45 mm and a thickness of 0.25 mm.

The disc is pressed between the mating surfaces of the cavities and fixed by a series of eight (M3) screws and bolts. Two threaded holes were present on the sidewall of the two cavities and served either for filling and evacuation of the cell with the liquid under test or for mounting two probes made up of a straight gold plated copper conductor which protruded 3 mm into the cylindrical cavity. During the microwave measurements the cell was placed inside a vacuum-tight vessel, within a thermostatted bath. The temperature of the cell was measured with a platinum resistance thermometer. Semi-rigid coaxial cables led from each probe via feedthroughs to a network analyzer (AGILENT 8719ES) which permitted measurements up to 13.5 GHz.

The instrumentation, materials and procedures used in the acoustic configuration are described elsewhere [4].

1. 2. Theory of microwave resonances in a cylindrical cavity

The experimental techniques based on the analysis of the frequency spectrum of cylindrical conducting cavities for a determination of their internal dimensions are well-established and have been used for many different applications [5, 6]. The normal modes in a right circular cylinder of diameter *D* and length *L* can be divided in TE- and TM-classes where the axis of reference *z* is along the cylinder axis. These modes are further specified in terms of three integers *m*, *n* and *p*, which are defined as the number of full-period variations of the different components of the electromagnetic field with respect to the angular, radial, and longitudinal coordinate. The normal mode fields are expressed in terms of the trigonometric and Bessel functions and the corresponding resonance frequencies, for the case of a cavity with perfectly conducting walls, are given by the equation:

$$f_{mnp}^2 = \left(\frac{c}{2p\sqrt{me}} \right)^2 \left(\frac{4c_{m,n}^2}{D^2} + \frac{p^2 p^2}{L^2} \right) \tag{1}$$

where *c*, m and e are respectively the speed of light, the magnetic permeability and the dielectric constant of the medium which fills the cavity, $c_{m,n}$ is the *n*-th root of $J_m(x) = 0$ for TM-modes (*m, p,* = 0, 1, 2, ...; *n* = 1, 2,...), or the n-th root of $J'_m(x) = 0$ (*m* = 0, 1, 2, ...; *n, p* = 1, 2,...) for TE-modes and $J_m(x)$ is the cylindrical Bessel function of order m. According to eq. (1) and to the selection rules reported above, the frequencies of the TM*mn*0 modes depend only on the cavity diameter *D* and not on its length *L* . All the other modes have resonance frequencies which depend both on *D* and *L*, so that a minimum of two modes is necessary for the determination of the length of a cylindrical cavity. However it should be noticed that from a metrological point of view the most appropriate choice for this purpose contemplates two *related* modes which differ only for the value of the index *p,* having equal values of the indexes *m* and *n*.

This being the case, we can consider the system:

$$\begin{cases} f^2_{mnp_1} = \left(\dfrac{c}{2p}\right)^2 \left(\dfrac{4c^2_{m,n}}{D^2} + \dfrac{p^2 p_1{}^2}{L^2}\right) & \text{(2a)} \\ f^2_{mnp_2} = \left(\dfrac{c}{2p}\right)^2 \left(\dfrac{4c^2_{m,n}}{D^2} + \dfrac{p^2 p_2{}^2}{L^2}\right) & \text{(2b)} \end{cases}$$

from which we obtain:

$$L^2 = \frac{c^2\left(p_2^2 - p_1^2\right)}{4}\left(f^2_{mnp_2} - f^2_{mnp_1}\right)^{-1} \tag{3}$$

As implied by the absence of *D* and by the squared difference dependence of *L* on $f^2_{mnp_i}$ in eq. (3), it is evident that possible systematic errors associated to the determination of the resonance frequencies of a couple of *related* modes would not affect the determination of *L* in the first order of approximation. Thus, apart from random errors associated with their measurement, the resonance frequencies of a couple of *related* modes are determined by exactly the same diameter and very nearly by the same length. As evidenced in Fig. 2, which shows the low frequency microwave spectrum of the two cavities which comprise the ultrasonic cell, the relative separation and position of modes of different symmetry depends on the value of the ratio D/L . The reported spectra were recorded with the two antennae mounted on the sidewall of the two cavities. This position does not allow excitation of the TM*mn*0 modes, whose frequency depends only on the cavity diameter and are of no interest for a determination of its length. The probes could also be alternatively mounted on the reflectors which form the basis of the cylindrical cavities. This position however did not allow detection of the whole class of the TE*mnp* modes. Positioning the antennae on the sidewall of the cavity then revealed two major advantages: i) inefficient detection of TM*mn*0 modes results in a better separation of low-frequency modes of different symmetry; ii) removal of the antennae from their ports for the acoustic measurements was possible without replacing or dismounting the reflectors, allowing a better reproducibility of the lengths alternatively sensed by the acoustic pulses and the microwave field. The results illustrated throughout the rest of this work were obtained with this configuration of the microwave probes. A casual inspection of Fig. 2 shows that, according to discussion above the TE111 and TE112 modes are the best candidates for a determination of the length L_1 and L_2 of the two cavities, in reason of their good isolation from other neighbouring modes.

1. 3. Data acquisition

Typically 101 frequencies spanning each mode under study each mode were recorded as the average of 10 scans with an IF bandwidth of 10 Hz. The network analyzer was configured to measure the four scattering coefficients $\mathbf{S}_{ij}$, however only the coefficient

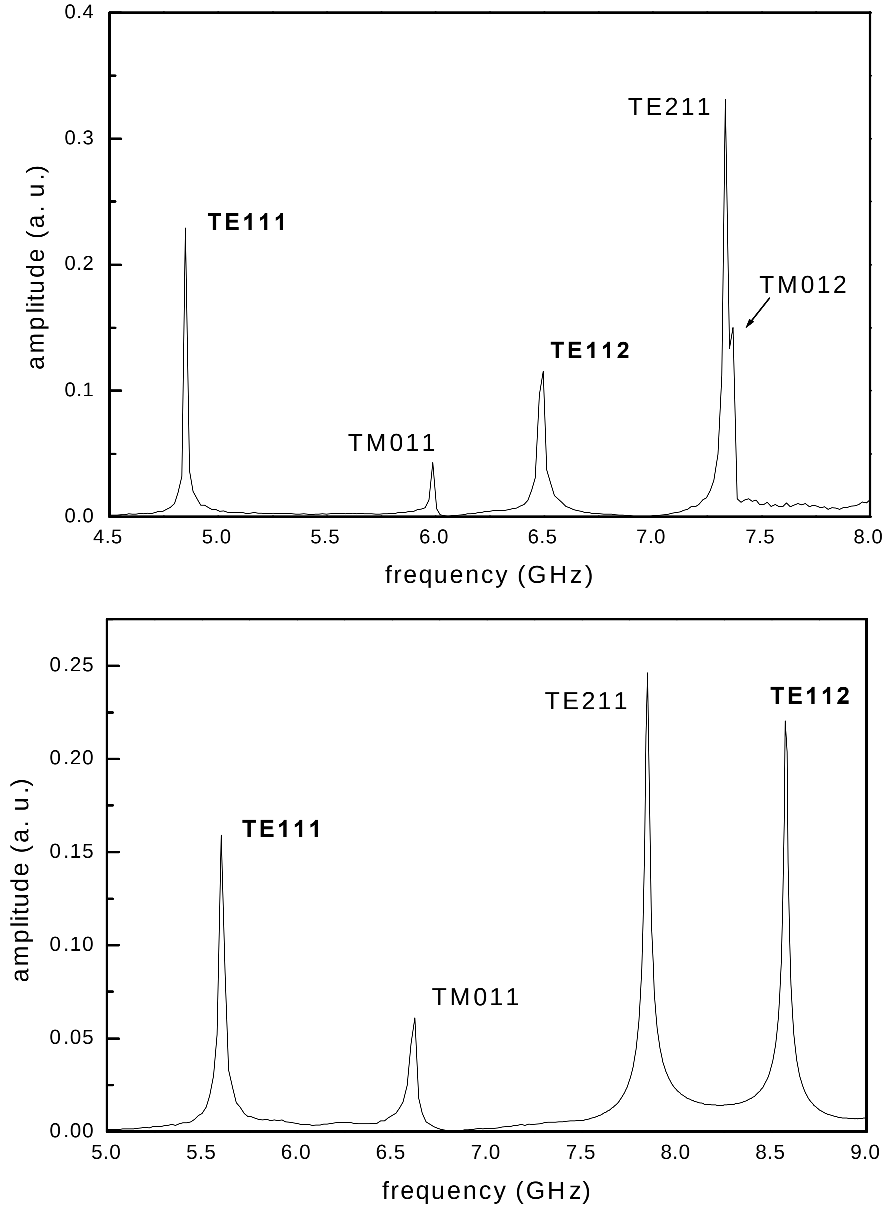

Fig. 2. Microwave spectrum of the two cylindrical cavities constituing the ultrasonic cell: Top; long cavity *D/L* = 0.70 Bottom: short cavity *D/L* = 1.05

$\mathbf{S}_{12}$ was utilised for the successive data analysis. The averaged real and imaginary parts of the signal were fitted to the complex lorentzian function of the frequency [7].

As an example of the extremely high level of precision of the fitting procedure and consequently of the resonance frequencies determination, Fig. 3 reports the results obtained in fitting the TE111 mode for the long cavity.

1. 4. Widths of microwave resonances

When the wall of the cavity has a finite conductivity s , the exponential decay of the electromagnetic field within the wall results in a contribution to the half-widths of the

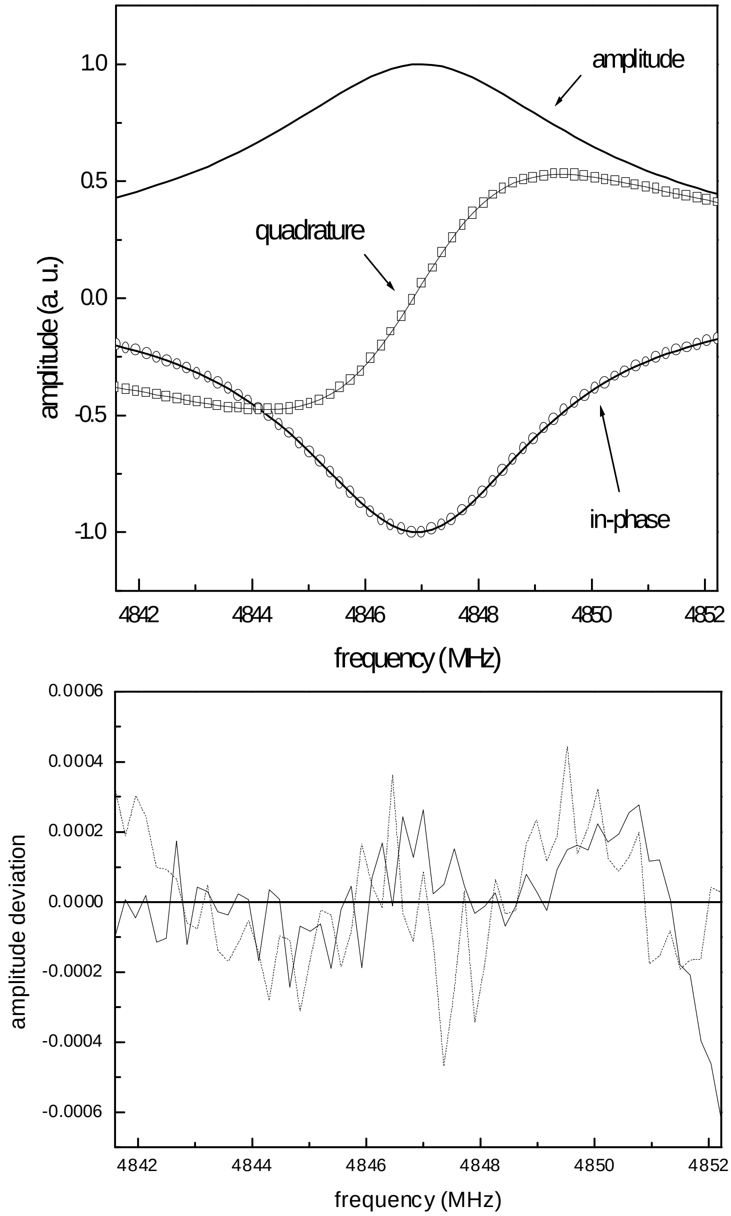

Fig. 3. Top: In-phase and quadraure signals detected by the network analyzer in correspondence of the TE111 mode in the longer cavity. Bottom: deviations of the detected signal from a fit with a complex lorentzian to the experimental data.

resonances and in an equal reduction of the frequencies. The magnitude of these effects is for TM- and TE- modes respectively [9]:

$$\frac{\Delta f_N + \mathrm{i} g_N}{f_N} = (-1+\mathrm{i}) \cdot d \cdot \left(\frac{1}{D} + \frac{1}{L} \right) \tag{4a}$$

$$\frac{\Delta f_N + \mathrm{i} g_N}{f_N} = (-1+\mathrm{i}) \cdot d \cdot \left(\frac{c_{mn}{}^2}{D\left(c_{mn}{}^2 - m^2\right)} + \frac{1}{L} - \frac{(pl)^2 D + 4 c_{mn}{}^2 L}{(pl)^2 D + 4 c_{mn}{}^2 L^2} \right) \tag{4b}$$

where N summarizes the set of three indexes (nmp), $\boldsymbol{F}_N = f_N + \mathrm{i}g_N$ is the complex resonance frequency of the mode under study (with g_N representing the halfwidth of the resonance curve which is determined by the various energy losses present in the system), and $\boldsymbol{d}$ is the penetration length given by

$$d = \left(p f_N m s\right)^{-0.5} \qquad (5)$$

In order to calculate g_N for the modes of interest we assumed $m = m_0$ the permeability of free space and we estimated the conductivity at 325 K as an average of the dc data reported in [8] for a variety of different stainless steel samples.

1. 5. Microwave Results

We made measurements in the vicinity of the modes TE111, TM011 and TE112 on a single isotherm near 325 K for the two cylindrical cavities comprising the ultrasonic cell.

Table I. Results of microwave measurements

mode	f_N(exp) (GHz)	g_N(exp) (MHz)	g_N(calc) (MHz)	$\Delta g_N/f_N$	s(fit)	$(\sigma f_N/f_N)$
			short cavity (D/L = 1.05) at T = 325.06 K			
TE111	5.6008838	4.628	1.156	$6.2\cdot10^{-4}$	$2.7\cdot10^{-4}$	$2.7\cdot10^{-7}$
TE112	8.5735303	11.113	1.448	$1.1\cdot10^{-3}$	$1.2\cdot10^{-3}$	$3.1\cdot10^{-6}$
TM011	6.6100239	5.215	1.788	$5.2\cdot10^{-4}$	$3.4\cdot10^{-4}$	$5.4\cdot10^{-7}$
			long cavity (D/L = 0.70) at T = 325.06 K			
TE111	4.8469746	2.583	0.997	$3.3\cdot10^{-4}$	$3.7\cdot10^{-4}$	$3.9\cdot10^{-7}$
TE112	6.4836382	3.840	1.070	$4.3\cdot10^{-4}$	$2.0\cdot10^{-4}$	$2.4\cdot10^{-7}$
TM011	5.9835830	3.159	1.406	$2.9\cdot10^{-4}$	$2.0\cdot10^{-4}$	$1.9\cdot10^{-7}$

The results are reported in Table I., where are listed: the experimental values of the resonance frequency f_N(exp) and halfwidth g_N(exp); the corresponding standard deviation s(fit) obtained from a fit procedure; the calculated value of the halfwdith g_N(calc) determined with eqs. (4a,b) and the data in [8]; the difference between experimental and calculated halfwidths scaled by the value of the resonance frequency

Dg_N/f_N ; the estimated relative random uncertainty associated to the frequency measurement (sf_N/f_N) calculated as $2(g_N/f_N)\,s(\text{fit})$.

When the isotherm at 325 K was completed and the results examined, it became evident, as shown from the data reported in Tab. I, a substantial disagreement between the experimental and calculated halfwidths for all the modes under study. A calculation of the expected resonance frequencies, made on the basis of the nominal dimensions at ambient temperature and the coefficient of thermal expansion, for the two cavities evidenced a systematic difference of similar entity. Following these observations we suspected the existence of a problem of overcoupling between the resonant cavity and the straight probe antennae used to excite and detect the microwave resonances. In order to test this hypothesis we dismounted one of the microwave probes of the short cavity and replaced it with a stainless steel plug whose front surface was machined to be flush with the interior surface of the cavity while reproducing its curvature. Successively we configured the network analyzer to measure the reflection coefficient $\mathbf{S}_{11}$ on a large frequency interval spanning the resonance curve of the mode TE112, and compared the results with those obtained with the two microwave probes and transmission lines connected to the cavity. This comparison, which is illustrated in Fig. 4, revealed a substantial decrease of the width of the resonance curve and a corresponding increase in the resonance frequency confirming the hypothesis discussed above.

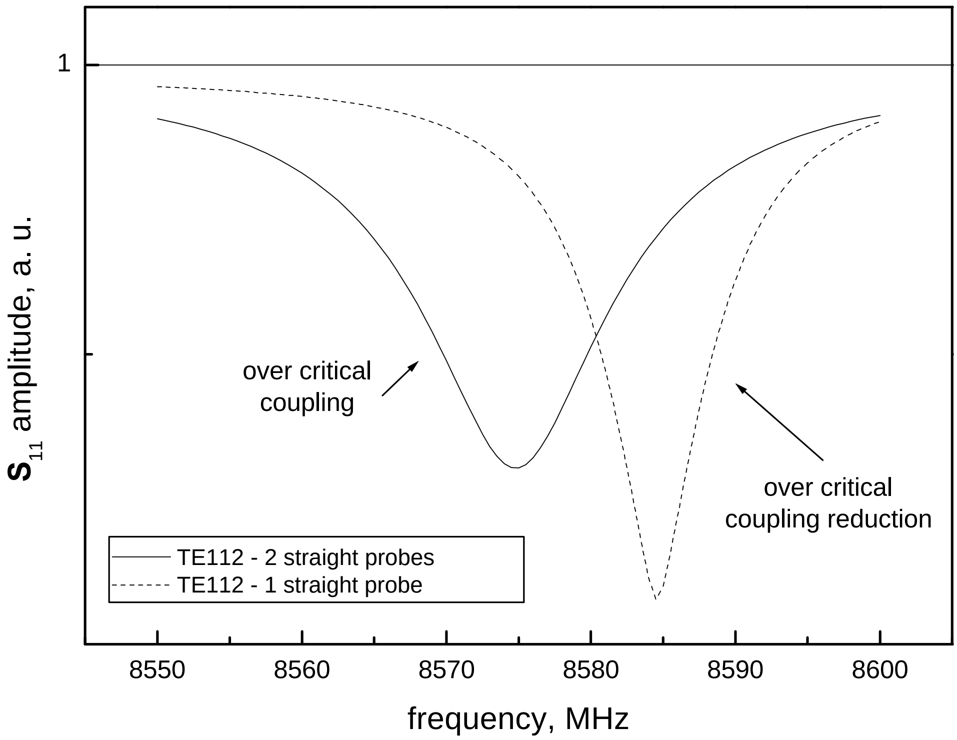

Fig. 4. Overcoupling effect on microwave resonances.

Though aware of this possible source of systematic error, we used the experimental halfwidths to correct the experimental frequencies of the modes TE111 and TE112 at 325 K for the two cavities and calculated the distances L_1 and L_2 between the transducer and the two reflectors and finally obtained the difference *D*L in the acoustic paths necessary to the calculation of the speed of sound from the propagation times obtained with the cell in the acoustic configuration as described below.

2. ACOUSTIC MEASUREMENTS

The double cavity cell design is the result of a compromise between the necessity of limited overall dimensions, which permit the insertion of the cell in our high pressure vessel, and the limited bandwidth of the network analyser. Since our aim was to check the feasibility of a high precision geometrical path determination, the ultrasonic cell design is not, in any way, the state-of-the-art in the field of acoustic sound speed measurements and could be subjected, in the future, to various and significant improvements.

To accomplish a first preliminary test of a sound speed measurements, water has been chosen in reason of the great amount of accurate literature data and its direct availability. The procedure was the following: as microwave measurements under vacuum had been carried out, antennae were dismounted and, without any other modification, the cell was placed in the high pressure vessel and thermostatted near 325 K. The vessel was filled with purified water, pressurised at 90 MPa, and pulse-echo measurements were effected at eleven pressure points regularly spaced between 90 MPa and 0.1 MPa.

The pulse-echo measurement is based on a double reflector method: the piezoelectric disk, which separates the two microwave cylindrical cavities during dimensional measurements, is used as an ultrasonic pulse source at its thickness mode resonance frequency. Repeated tone bursts are delivered by a function generator to the piezo disk, and echoes coming from reflectors (the other end-faces of the two cylindrical microwave cavities) are sampled by a digital oscilloscope. Time delays analysis is accomplished as explained in [4]; from the combined results of the echoes time of flight and the dimensional measurements, speed of sound was finally obtained.

From an acoustical point of view, the cylindrical double cavity, with the piston-like source placed at its base, occupying the entire section of the tube, behaves as a wave guide for acoustic wave propagation. In this experimental configuration, sound speed measurements, even if not affected by diffraction [10], are subjected to adsorption and dispersion effects [11]. A sound wave propagating along a tube interacts with the side wall and undergoes thermal and viscosity losses, so presenting a modified phase velocity, with respect to the free-field condition, satisfying the relation

$$u = u_* \left[1 + \left(\frac{u_*}{w} \right) a_{KH} \right], \tag{6}$$

where u is the free-field phase velocity, u_* is the guided-wave phase velocity, w is the angular frequency and a_{KH} is the Kirchhoff-Helmholtz adsorption coefficient, which depends on the transport properties of the medium (b is the radius of the circular tube,

k the thermal conductivity, h the viscosity, r the density of the fluid, C_P and C_V the specific heats at constant pressure and constant volume respectively, $g = C_P / C_V$):

$$a_{KH} = \frac{w}{2ub}\left[d_{sv} + (g-1)d_{th}\right], \tag{7}$$

$$d_{th} = \sqrt{\frac{2k}{rC_P w}}, \tag{8}$$

$$d_{sv} = \sqrt{\frac{2h}{rw}}. \tag{9}$$

This analysis, applied to our experimental configuration, leads to a correction in the order of 5 ppm, completely negligible with respect to the other major sources of uncertainty. Dispersion effects, on the other hand, could have non negligible influence on the acoustic time of flight determination . The carrier frequency of the acoustic pulses, being well above the cut-off frequency of the wave-guide (which for the present experimental setup, is located at about 45 kHz), is compatible with higher modes of wave transmission, other then the ordinary plane-wave longitudinal mode.

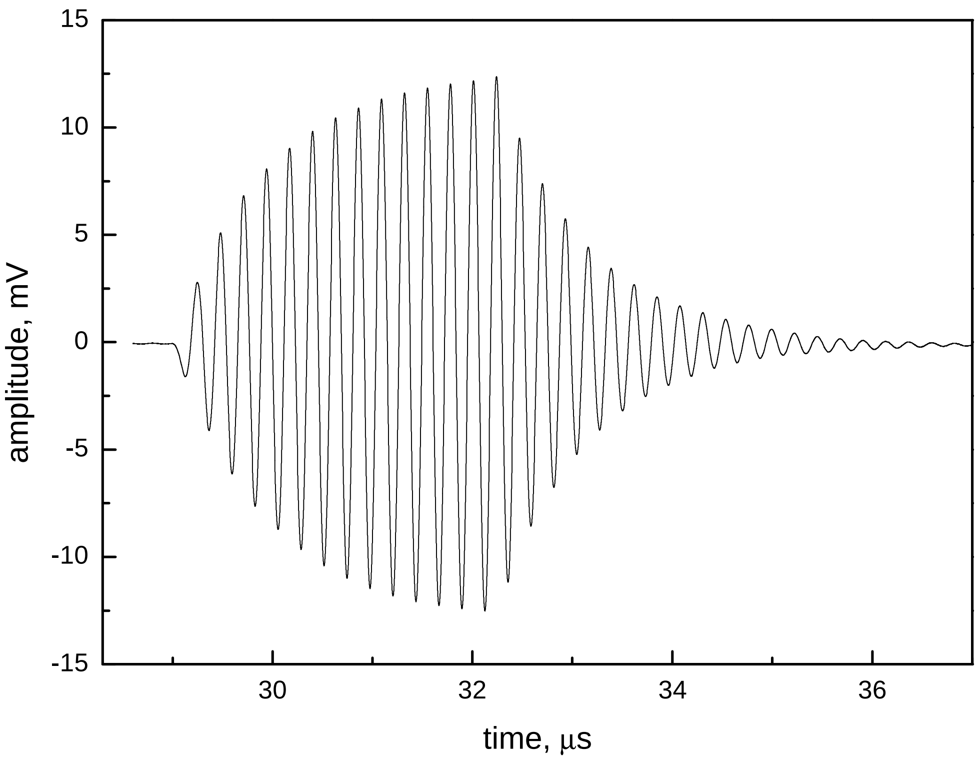

Fig. 5. Acoustic pulse emitted by piezoceramic transducer, as recorded by a probe hydrophone.

Those higher modes have phase velocities greater than the sound speed in free space which depend on the frequency of the signal

$$u_{phase} = \frac{u}{\sqrt{1-(uc_n/w)}}, \tag{10}$$

where w is the angular frequency of the signal, u is the sound speed in free field and c_n a numerical constant depending on the symmetry of the mode and on boundary conditions The corresponding group velocities, the only quantities accessible by experiment, are lower than the free field speed:

$$u_g = u\sqrt{1-(uc_n/w)} = \left(\frac{u}{u_{phase}}\right)u. \tag{11}$$

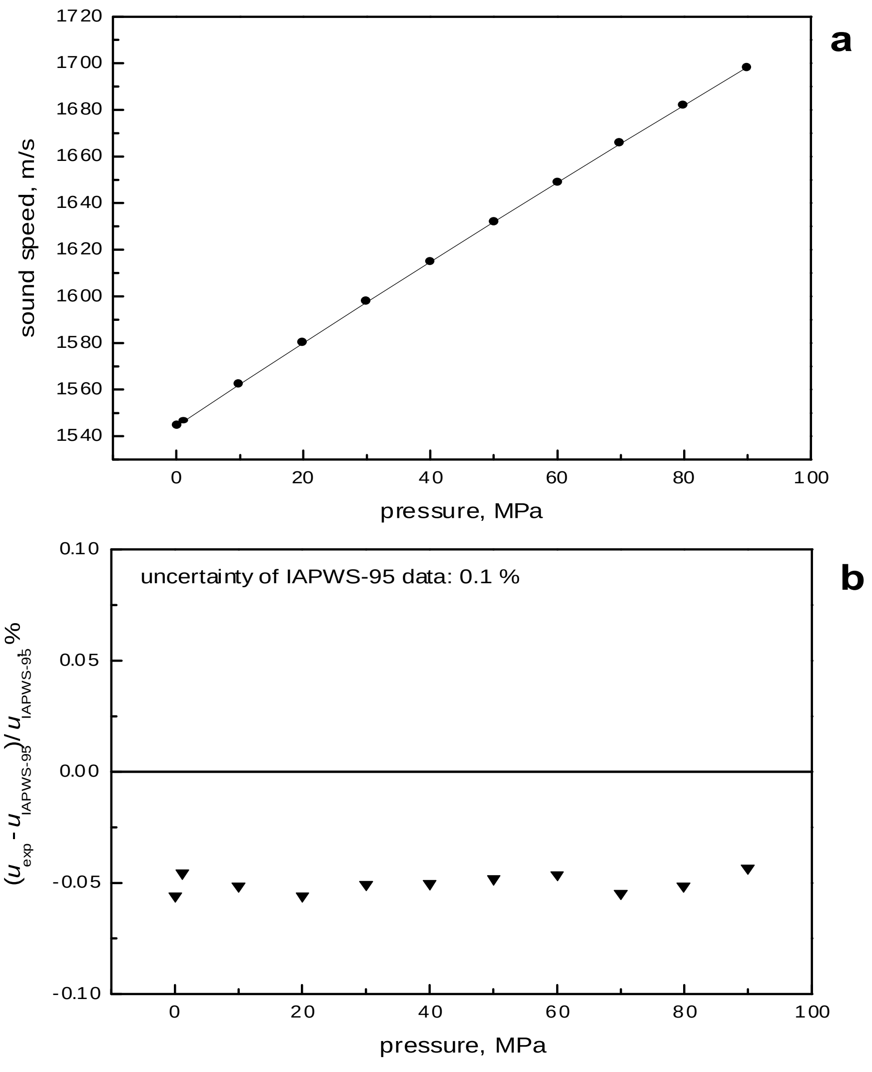

Fig. 6. a): Experimental speed of sound in water at 325 K; b) Relative deviations from IAPWS-95 formulation.

If these non-longitudinal modes are excited by the source, the shape of acoustic pulses is distorted by dispersion and the measured sound speed does not coincide with the thermodynamic sound speed. In order to check possible dispersion effects introduced by the cavities in our pulse echo experiments, we analysed the acoustic signal emitted by the transducer by means of a hydrophone probe. A computer recorded trace from the hydrophone is shown in Fig. 5. Apparently, the signal is not significantly influenced by wave-guide induced distortions, confirming that the transducer undergoes a nearly piston-like motion as a consequence of electrical excitation.

3. PRELIMINARY RESULTS AND CONCLUSIONS

Data from pulse-echo measurements, in the form time of flight determinations, have been combined with microwave dimensional measures for a single isotherm at 325 K. The resulting sound speed as a function of pressure is shown in Fig. 6a. The relative precision of dimensional measurements, as calculated from standard deviations in microwave resonance frequencies determinations, is about $1{\cdot}10^{-5}$; the overall uncertainty in speed of sound evaluation, as illustrated in [4], results to be about 0.05% for the experimental apparatus used in these preliminary tests. A comparison with available data drawn from IAPWS-95 formulation is displayed in Fig. 6b. The obtained data appear to be affected by a low random noise, but a noticeable systematic deviation is present. We attribute this lack of accuracy to the imperfect acoustic performance of the ultrasonic cell, due to the poor efficiency of the 4 mm thick reflectors. Ultrasonic pulses propagate in the solid medium and are back-reflected by the outer surface towards the piezoelectric disk, interfering with the main acoustic echoes. This problem would be overcome with a reshaping of the back surface of the reflectors, in order to improve their sound adsorption and dispersion characteristics.

REFERENCES

1. P. J. Kortbeek, M. J. P. Muringer, N. J. Trappeniers and S. N. Biswas, *Rev. Sci. Instrum*, **56**:1269, (1985).
2. S. J. Ball and J.P.M. Trusler, *Int. J. Thermophys.* **22**:427 (2001).
3. W. Wagner and A. Pruss, *J. Phys. Chem. Ref. Data* **31**:387 (2002).
4. G. Benedetto, R. M. Gavioso, P. A. Giuliano Albo, S. Lago, D. Madonna Ripa and R. Spagnolo, in *Speed of Sound of Pure Water at Temperatures between 274 and 394 K and Pressures up to 90 MPa*, paper presented at the Fifteenth Symposium on Thermophysical Properties, (Boulder, 2003)
5. K. D. Froome and L. Essen, in *Velocity of light and radio waves*, (Academic Press, New York, 1969).
6. W. J. Radcliffe, J. C. Gallop, *J. Phys. E: Sci. Instrum.*, **14**: 461 (1981).

7. M. R. Moldover, S. J. Boyes, C. W. Meyer and A. R. H. Goodwin*, J. Res. Natl. Inst. Stand. Technol.*, **104**, 11 (1999).
8. A. F. Clark, G. E. Childs and G. H. Wallace, in Advances in Criogenic Engineering, Vol. 15, K. D. Timmerhaus, ed. (Plenum Press, New York and London, 1970), pp. 85-90.
9. C. J. Montgomery, in *Technique of microwave measurements*, (Mc Graw-Hill, New York and London, 1947), pp.300-302.
10. P.M. Trusler, in *Physical Acoustics and Metrology of Fluids*, (The Adam Hilger Series On Measurement Science and Technology, 1991).
11. P. M. Morse, K. U. Ingard, in *Theoretical Acoustic*, pp 492-501, (Princeton University Press, 1986)